

**SOLID ELECTROLYTE COMPOSITION,  
METHOD FOR MANUFACTURING THE  
SAME, AND ELECTRODE SHEET FOR  
BATTERY AND ALL-SOLID-STATE  
SECONDARY BATTERY IN WHICH SOLID  
ELECTROLYTE COMPOSITION IS USED**

**CROSS-REFERENCE TO RELATED  
APPLICATIONS**

**[0001]** This application is a Continuation of PCT International Application No. PCT/JP2015/054368 filed on Feb. 18, 2015, which claims priority under 35 U.S.C. §119 (a) to Japanese Patent Application No. JP2014-033286 filed in Japan on Feb. 24, 2014.

**[0002]** Each of the above applications is hereby expressly incorporated by reference, in its entirety, into the present application.

**BACKGROUND OF THE INVENTION**

**[0003]** 1. Field of the Invention

**[0004]** The present invention relates to a solid electrolyte composition, a method for manufacturing the same, and an electrode sheet for a battery and an all-solid-state secondary battery in which the solid electrolyte composition is used.

**[0005]** 2. Description of the Related Art

**[0006]** An electrolyte solution is used in a lithium ion battery which is widely used currently in many cases. There has been an attempt to cause all configuration materials to be solid by substituting the electrolyte solution with a solid electrolyte. Above all, the advantages of the technique of using an inorganic solid electrolyte are reliability at the time of usage and stability. A combustible material such as a carbonate-based solvent is applied as a medium of the electrolyte solution which is used in the lithium ion secondary battery. Various measures are employed, but an additional measurement to be performed when a battery is overcharged is desired. An all-solid-state secondary battery formed of an inorganic compound that can cause an electrolyte to be incombustible is regarded as fundamental solving means thereof.

**[0007]** Another advantage of the all-solid-state secondary battery is that a high energy density is suitably achieved by stacking electrodes. Specifically, the all-solid-state secondary battery can be a battery having a structure in which electrodes and electrolytes are directly arranged side by side to be serialized. At this point, a metal package that seals battery cells and a copper wire or a bus bar that connects battery cells can be omitted, and thus an energy density of the battery can be greatly increased. It is advantageous that compatibility with a positive electrode material in which a potential can be enhanced to a high level is good.

**[0008]** According to the respective advantages as described above, the development of the all-solid-state secondary battery as a next-generation lithium ion secondary battery is vigorously advanced (see NEDO: New Energy and Industrial Technology Development Organization, Fuel Cells-Hydrogen Technology Development Field, Electricity Storage Technology Development Division “NEDO 2008 Roadmap for the Development of Next Generation Automotive Battery Technology” (June 2009)). In the all-solid-state secondary battery, an inorganic solid electrolyte layer is particularly a member that does not exist in a liquid-type battery or a polymer-type battery, and the development

thereof is emphasized. This solid electrolyte layer is generally formed by heating and pressurizing an electrolyte material applied thereto together with a binder. Accordingly, the adhesion state between the solid electrolyte layers is replaced from a point contact to a surface contact, particle boundary resistance is decreased, and impedance is decreased. A forming example of an all-solid-state lithium battery to which this step is employed is known (see JP3198828B). There is an example in which an average particle diameter (number average particle diameter) of the solid electrolyte particles thereof or the distribution thereof is caused to have a specific scope (see WO2011/105574A). Accordingly, it is considered that a slurry composition having favorable dispersibility and coatability can be obtained.

**SUMMARY OF THE INVENTION**

**[0009]** According to the technique disclosed in WO2011/105574A, suitability of manufacturing may be improved as described above. However, if recently increasing demand on high performances required in the all-solid-state secondary battery is considered, development of techniques that can satisfy higher levels is required.

**[0010]** Therefore, the invention has an object of providing a solid electrolyte composition that can realize improved ion conductivity in an all-solid-state secondary battery and a method for manufacturing the same, and an electrode sheet for a battery and an all-solid-state secondary battery in which the solid electrolyte composition is used.

**[0011]** The problems are solved by the means below.

**[0012]** [1] A solid electrolyte composition comprising: inorganic solid electrolyte particles exhibiting at least two peaks in accumulative particle size distribution which is measured with a dynamic light scattering-type particle diameter distribution measuring device.

**[0013]** [2] The solid electrolyte composition according to 1, in which, among the two or more peaks, a peak (Pa) of a maximum particle diameter is in the particle diameter range of 2  $\mu\text{m}$  to 0.4  $\mu\text{m}$  and a peak (Pb) of a minimum particle diameter is in the range of 1.5  $\mu\text{m}$  to 0.1  $\mu\text{m}$ , and a relationship between the peak (Pa) of the maximum particle diameter and the peak (Pb) of the minimum particle diameter satisfies Expression (1) below.

$$0.05 \leq P_b/P_a \leq 0.75 \quad (1)$$

**[0014]** [3] The solid electrolyte composition according to 1 or 2, in which the inorganic solid electrolyte particles include inorganic solid electrolyte particles A having an average particle diameter (da) of 2  $\mu\text{m}$  to 0.4  $\mu\text{m}$  and inorganic solid electrolyte particles B having an average particle diameter (db) of 1.5  $\mu\text{m}$  to 0.1  $\mu\text{m}$ , and Expression (2) below is satisfied.

$$0.05 \leq d_b/d_a \leq 0.75 \quad (2)$$

**[0015]** [4] The solid electrolyte composition according to any one of 1 to 3, in which, with respect to the accumulative particle size distribution measured with the dynamic light scattering-type particle diameter distribution measuring device, when respective peaks are assumed to follow log-normal distribution and the waveform is separated by a nonlinear least square method, an accumulative 90% particle diameter (Pa90) of a peak (Pa) of a maximum particle diameter is 3.4  $\mu\text{m}$  to 0.7  $\mu\text{m}$ , and an accumulative 90%